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RESEARCH LETTER

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Key Points:

- Measurements of all the major synthetic greenhouse gases have been compiled
- These measurements have been used to infer recent global emissions trends
- Based on these trends, future emissions scenarios have been investigated

Supporting Information:

- Readme
- Synthetic GHG scenario development
- Synthetic GHG inverse method
- C3F8 emissions
- C2F6 emissions
- SO₂F₂ emissions
- SF₆ emissions
- NF₃ emissions
- HFC365mfc emissions
- HFC32 emissions
- HFC245fa emissions
- HFC23 emissions
- HFC227ea emissions
- HFC152a emissions
- HFC143a emissions
- HFC134a emissions
- HFC125 emissions
- HCFC22 emissions
- HCFC142b emissions
- HCFC141b emissions
- CH₃CCl₃ emissions
- CFC12 emissions
- CFC115 emissions
- CFC114 emissions
- CFC113 emissions
- CFC11 emissions
- CF₄ emissions
- CCl₄ emissions

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Recent and future trends in synthetic greenhouse gas radiative forcing

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Abstract Atmospheric measurements show that emissions of hydrofluorocarbons (HFCs) and hydrochlorofluorocarbons are now the primary drivers of the positive growth in synthetic greenhouse gas (SGHG) radiative forcing. We infer recent SGHG emissions and examine the impact of future emissions scenarios, with a particular focus on proposals to reduce HFC use under the Montreal Protocol. If these proposals are implemented, overall SGHG radiative forcing could peak at around 355 mW m⁻² in 2020, before declining by approximately 26% by 2050, despite continued growth of fully fluorinated greenhouse gas emissions. Compared to “no HFC policy” projections, this amounts to a reduction in radiative forcing of between 50 and 240 mW m⁻² by 2050 or a cumulative emissions saving equivalent to 0.5 to 2.8 years of CO₂ emissions at current levels. However, more complete reporting of global HFC emissions is required, as less than half of global emissions are currently accounted for.

1. Monitoring Global Trends in Synthetic Greenhouse Gases

In 2012, the major long-lived synthetic greenhouse gases (gases with no significant natural sources and lifetimes of at least 1 year) were responsible for 350 ± 10 mW m⁻² of direct radiative forcing (RF), 19% as large as the increase in RF due to CO₂ since the preindustrial era [Hall et al., 2012] (note that the uncertainty in SGHG RF excludes radiative transfer assumptions, which are estimated to be of the order of 10% [Forster et al., 2007]). This group consists of a few tens of compounds, which we break down into the following groups (in the order of their current contribution to RF): chlorofluorocarbons (CFCs) and other strongly ozone depleting substances (ODS) (which include CCl₄ and CH₃CCl₃ here), hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), fully fluorinated GHGs (FFGHGs) (consisting of perfluorocarbons, SF₆, and NF₃), and SO₂F₂. The CFCs and their replacements, the HCFCs and HFCs, are primarily used in refrigeration, air conditioning, and foam blowing [e.g., Montzka et al., 2011]. FFGHGs are emitted during aluminum manufacture (primarily CF₄) and are used in a range of applications such as electrical insulation (e.g., SF₆) or semiconductor manufacture (e.g., NF₃ and C₂F₆) [Weiss et al., 2008; Mühle et al., 2010; Rigby et al., 2010; Arnold et al., 2013]. Some gases, such as HFC-23 (CHF₃), have little practical use but are released to the atmosphere as unwanted by-products during certain industrial processes [Miller et al., 2010; Miller and Kuijpers, 2011]. Despite being present in the atmosphere at levels of only a few hundred parts per trillion or less, synthetic greenhouse gases (SGHGs) have a significant climate impact because of their very high radiative efficiencies and, in many cases, very long lifetimes (tens to thousands of years) [Ravishankara et al., 1993; Forster et al., 2007].

Here we examine recent trends in 25 of the most abundant SGHGs measured by the Advanced Global Atmospheric Gases Experiment (AGAGE) [Prinn et al., 2000]: CFC-11, CFC-12, CFC-113, CFC-114, CFC-115, CCl₄, CH₃CCl₃, HCFC-22, HCFC-141b, HCFC-142b, HFC-23, HFC-32, HFC-125, HFC-134a, HFC-143a, HFC-152a, HFC-227ea, HFC-245fa, HFC-365mfc, CF₄, C₂F₆, C₃F₈, SF₆, NF₃, and SO₂F₂. There are some SGHG with an RF known to be higher than some of the more minor members of this list, which are not included here because AGAGE measurements are not yet available for all or part of the time period investigated. Examples of these compounds include c-C₄F₈ [Oram et al., 2012], some halons [Fraser et al., 1999], and some minor CFCs and HCFCs [e.g., Culbertson et al., 2004; Maione et al., 2013]. Measurements have also been made of SGHGs

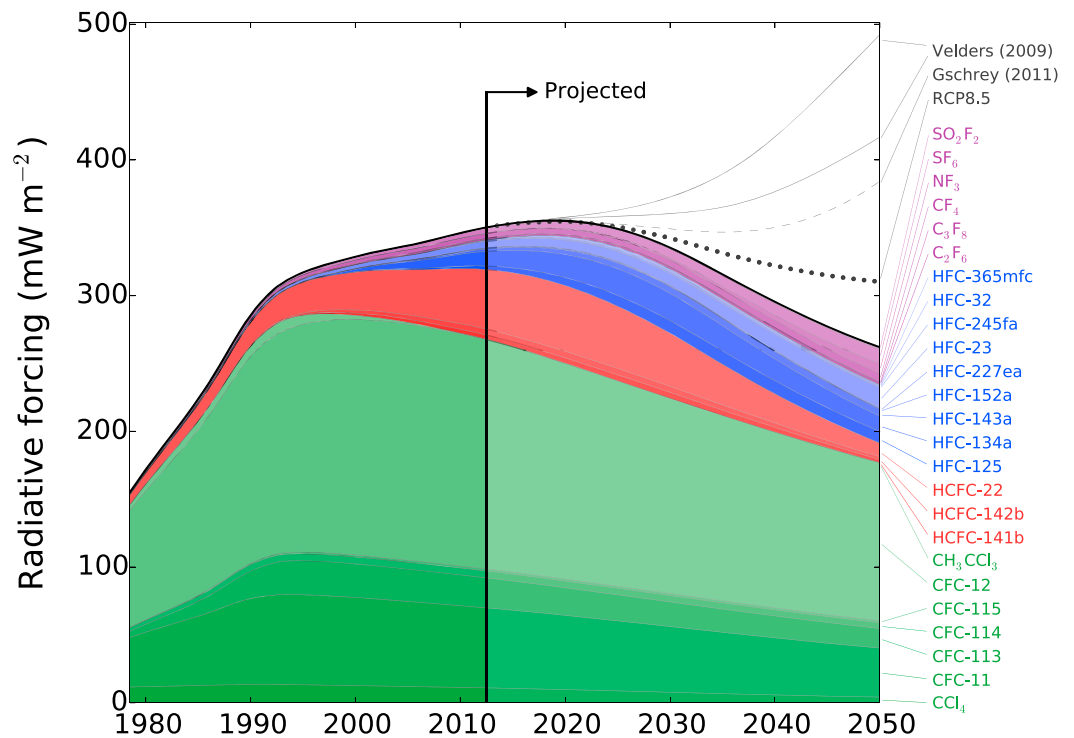


Figure 1. Global radiative forcing due to long-lived synthetic greenhouse gases, derived from AGAGE observations from 1980 to 2012 (darkly shaded areas). The more lightly shaded areas show the predicted radiative forcing due to each SGHG from 2013 until 2050, assuming that recent plans to phase down HFCs are implemented, existing Montreal Protocol obligations are met, and emissions of FFGHG show moderate growth. The solid, dotted, and dashed lines show SGHG radiative forcing under three previously published HFC scenarios that do not involve major emissions abatement measures.

that exert a smaller influence on global RF than the 25 investigated here [e.g., *Straume et al., 1998; Sturges et al., 2000; Simmonds et al., 2002; Vollmer et al., 2011; Ivy et al., 2012a, 2012b*]. We estimate that the combined RF due to these omitted species was of the order of 1% of the overall SGHG RF in 2012.

Since 1978, AGAGE and its previous incarnations as the Atmospheric Lifetimes Experiment and Global Atmospheric Gases Experiment have charted the trends in SGHG from remote stations, chosen to observe “background” air masses, using gas chromatography-electron capture detection and gas chromatography-mass spectrometry techniques [*Simmonds et al., 1995; Prinn et al., 2000; Miller et al., 2008; Arnold et al., 2012*]. The data used in this paper have been compiled from both in situ AGAGE observations and new and previously published measurements of archived air samples from Cape Grim, Tasmania and several northern hemispheric locations [*O’Doherty et al., 2004, 2009; Miller et al., 2010; Mühle et al., 2010; Rigby et al., 2010; Vollmer et al., 2011; Saikawa et al., 2012; Arnold et al., 2013*]. We have assimilated these observations into a model of atmospheric chemistry and transport to determine global trends and infer emissions of the entire suite of SGHG, using the methodology outlined in *Rigby et al. [2011, 2013]* and in the supporting information. Atmospheric lifetimes in the model are based on the recent evaluation carried out under the Stratospheric-troposphere Processes and their Role in Climate (SPARC) Lifetime of Halogen Source Gases report [*SPARC, 2013*].

2. Recent Trends

Global RF due to SGHGs from 1978 to 2012, calculated using the assimilated suite of AGAGE observations, is shown in Figure 1. Overall RF has been dominated by the CFCs from the beginning of this record. However, since the international ratification of the Montreal Protocol in 1987 and subsequent strengthening through amendments during the 1990s, the reported use of CFCs has declined dramatically. This has led to a 7% reduction in overall global CFC and other ODS RF since the peak in 1996, making the Montreal Protocol a relatively effective piece of global climate legislation

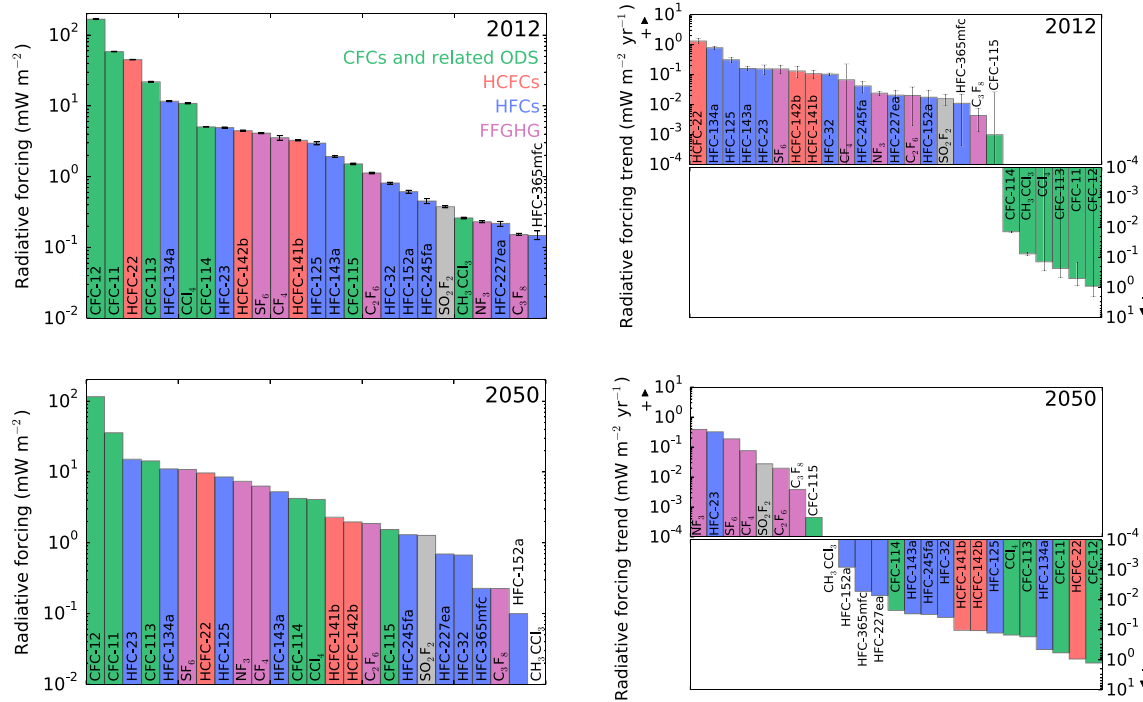


Figure 2. (left) Radiative forcing and (right) 5 year averaged trend observed in the (top) years preceding 2012 and (bottom) predicted for 2050, assuming moderate FFGHG emissions growth trajectories and Montreal Protocol HFC, CFC, and HCFC phase-down. One-sigma uncertainties in radiative forcing due to the AGAGE assimilation are shown in Figure 2, top as error bars.

(as noted by Molina *et al.* [2009], Velders *et al.* [2012], and Estrada *et al.* [2013]). In response to the phaseout of CFCs, use of HCFCs and HFCs has increased in similar applications. AGAGE observations show a rapid growth in concentrations of HCFCs and HFCs over the last two decades, such that in 2012, HCFCs and HFCs were responsible for 15% and 6% of the SGHG radiative forcing, respectively. The FFGHGs have made a more minor contribution to 2012 radiative forcing, currently at 3% of overall SGHG RF. However, their long lifetimes mean that their radiative impact will only increase in the coming decades, even if emissions drop to relatively low levels.

Analysis of the contributions of individual species to global RF reveals that CFC-12 remains the most important SGHG (Figure 2), with a RF of $168 \pm 2 \text{ mW m}^{-2}$ in 2012, making it responsible for $48 \pm 3\%$ of the SGHG RF. However, it is also the species whose RF is declining the most quickly, with a trend of $-0.9 \pm 1 \text{ mW m}^{-2} \text{ yr}^{-1}$ (averaged over the 5 years up to 2012). In fact, all of the CFCs and ODSs examined here, except for CFC-115 (which has a lifetime of around 1020 years) [SPARC, 2013], show significantly negative RF growth trends in recent years. In contrast, every other species is increasing its contribution to global RF, with rates that vary by almost 4 orders of magnitude. The major non-CFC contributor to both SGHG RF and RF growth is HCFC-22, which was adding $1.3 \pm 0.3 \text{ mW m}^{-2} \text{ yr}^{-1}$ to its contribution of $45 \pm 2 \text{ mW m}^{-2}$ in recent years. Of the HFCs, HFC-134a currently makes a smaller contribution to overall RF than HCFC-22 ($12 \pm 0.2 \text{ mW m}^{-2}$), but its RF trend is of a similar order of magnitude, at $0.8 \pm 0.2 \text{ mW m}^{-2} \text{ yr}^{-1}$. Despite being relatively minor contributors to global RF, some HFCs are growing particularly quickly. For example, HFC-125 and -143a are the thirteenth and fourteenth most important contributors to absolute SGHG RF, but are growing at the third and fourth highest rates, respectively. Of the FFGHGs, SF₆ is currently the most significant, both in terms of RF and RF growth.

3. Global HFC and PFC Emissions Under-Reporting

To investigate the drivers of these trends, we compared the emissions inferred from AGAGE observations to available reports to the United Nations Framework Convention on Climate Change

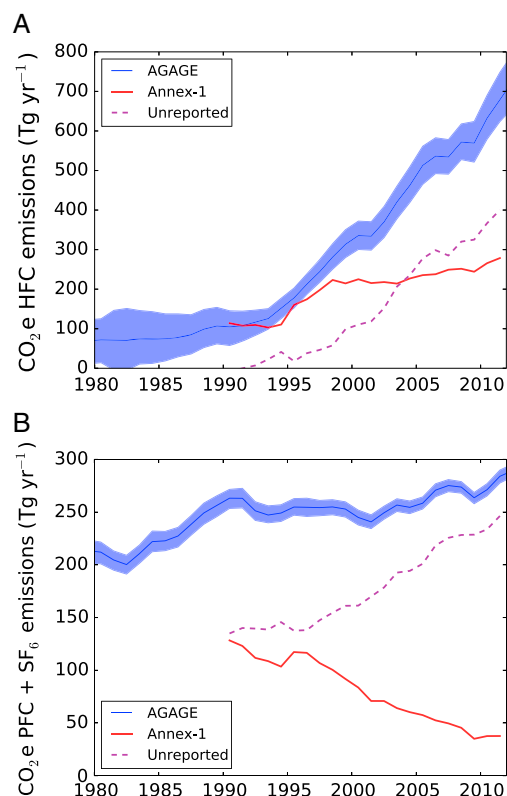


Figure 3. (a) HFC and (b) PFC and SF₆ CO₂-equivalent emissions derived from AGAGE observations (blue line and shading, representing 1-sigma uncertainties) compared to reported emissions from Annex 1 countries (red). Unreported emissions (AGAGE minus reported emissions) are shown in purple.

discrepancy has increased significantly in recent years, with inferred global emissions growing at the same time as reported emissions have declined. As a result, over 90% of combined PFC and SF₆ emissions were unreported in 2012.

These comparisons highlight at least two potential issues with current emissions reporting. First, incomplete global reporting is likely to leave much of the world's emissions unaccounted for. For example, recent observations from East Asia indicate significant emissions of HFCs and FGHGs from non-Annex 1 countries in the region, notably China [Kim *et al.*, 2010; Yao *et al.*, 2012]. Additionally, the growing discrepancy between reported and inferred emissions for both groups of compounds could also be indicative of growth in emissions from growing nonreporting economies. Second, the emissions reports may themselves be subject to inaccuracies. For example, recent regional emissions estimates inferred from atmospheric measurements suggest that SF₆ may be under-reported by Annex 1 countries [Levin *et al.*, 2010; Rigby *et al.*, 2010]. Similar discrepancies in other gases could be partly responsible for the overall difference between the reported and derived emissions.

In the following section, we explore possible future RF trajectories for each group of SGHG, particularly in light of potential changes to the Montreal Protocol. The emissions derived above serve as a 2012 “baseline” from which projections to 2050 can now be made.

4. Future RF Trajectories and Possible HFC Phase-Down

Owing to their potential to deplete stratospheric ozone, the HCFCs are currently being phased-down in developed countries under the Montreal Protocol, and will begin phase-down in developing countries in 2015. By 2030, almost all HCFC consumption should be halted. HCFCs, which have lifetimes of around 10 years, are removed from the atmosphere much more quickly than CFCs, which have lifetimes of around 50

(UNFCCC). So-called “Annex 1” countries (including the USA, EU members, and many other developed countries) have been required to report emissions of HFCs, perfluorocarbons (PFCs), and SF₆ based on “bottom-up” accounting methodologies from 1990 onward. For the HFCs, these reports initially showed a rapid rise in emissions, resulting in more than a doubling of the global release rate in the 1990s, before emissions growth slowed significantly from around 1998 onward (Figure 3). Global emissions inferred from AGAGE observations show relatively good agreement with those reported in the early 1990s. However, emissions have continued to rapidly increase after reported emissions growth slowed, to such an extent that less than half of the world's HFC emissions were unaccounted for in 2011, the last year for which emissions reports were available (680 ± 70 Tg-CO₂e yr⁻¹ compared to 279 Tg-CO₂e yr⁻¹ reported).

Compared to the dramatic growth in HFC emissions, aggregated emissions of the PFCs and SF₆ have grown more moderately between 1980 and 2012. Recent studies have noted that SF₆ emissions increased significantly during this period [Rigby *et al.*, 2010], whereas emissions of the major PFC, CF₄, have declined [Mühle *et al.*, 2010]. In contrast to the HFCs, emissions of PFCs and SF₆ have never been fully accounted for by reported release rates from Annex 1 countries. In 1990, more than half of the CO₂-equivalent global emissions were unreported and this

Table 1. Emissions Scenarios for Each Group of SGHG and Resulting Range in Predicted Radiative Forcing (Including the 1-Sigma Range in Atmospheric Lifetimes as Estimated by SPARC [2013])^a

Group	Scenarios	2050 RF Range (mW m^{-2})
CFCs	Daniel et al. [2011] RCP3-PD, 4.5, 6, and 8.5 ^b	169–187
HCFCs	Daniel et al. [2011] RCP3-PD RCP4.5, 6, and 8.5 ^b	13–41
HFCs (excluding HFC-23)	This work (US EPA phase-down) RCP3-PD RCP4.5 RCP6 RCP8.5 <i>Velders et al. [2009]</i> ^c <i>Gschrey et al. [2011]</i>	25–281
HFC-23	Miller and Kuijpers [2011] ^d RCP3-PD RCP4.5 RCP6 RCP8.5	4–15
FFGHGs	RCP3-PD RCP4.5 RCP6 RCP8.5 <i>Gschrey et al. [2011]</i>	20–28

^aProjections in bold were used in the “HFC phase-down” scenario.

^bEmissions in each of these scenarios are identical.

^cBoth “high” and “low” emissions growth scenarios included.

^dThe Miller and Kuijpers [2011] “Reference case” scenario was used in our HFC phase-down projections, and their “less mitigation” and “best practices” scenarios were included in the sensitivity study.

to hundreds of years. Therefore, the phase-down of these compounds has the potential to provide relatively rapid global climate benefits. Using emissions projections of CFC/ODS and HCFC from 2012 to 2050 that are consistent with the atmospheric mole fractions in Daniel et al. [2011], we estimated their future RF. We find that RF due to CFCs/ODSs should decline steadily in the coming decades as lingering emissions gradually decline and the gases are photochemically removed from the atmosphere (Figure 1). By 2050, CFC/ODS RF could be 34% lower than in 2012 and 41% lower than the maximum (which occurred in 1996). Despite their comparatively delayed phase-down schedule, the HCFCs show a more dramatic 73% reduction in RF by 2050 (compared to 2012), reflecting their more rapid removal from the atmosphere than CFCs. Under this scenario, HCFC RF is projected to peak in 2020 at 57 mW m^{-2} .

No global legislation currently exists to regulate the use of HFCs or FFGHG. However, recent proposals have been made to phase down global HFC consumption through the Montreal Protocol. Using the US Environmental Protection Agency’s proposed consumption phase-down schedule [Environmental Protection Agency (EPA), 2013] and assumptions about release rates from HFC “banks” (following Velders et al. [2009]), we have developed a projection of HFC emissions to 2050 (see supporting information for detailed scenario development). The radiative forcing predicted using this emissions trajectory is shown in Figure 1. HFC-23, which is a potent greenhouse gas, and unwanted by-product of HCFC-22 production, is not included in the US EPA proposals. The RF projection in Figure 1 assumes that HFC-23 emissions follow the “reference” scenario of Miller and Kuijpers [2011]. Estimates of future radiative forcing due to FFGHGs are based on the Representative Concentration Pathway (RCP) scenarios [Fujino et al., 2006; Smith and Wigley, 2006; Clarke et al., 2007; Riahi et al., 2007; van Vuuren et al., 2007; Meinshausen et al., 2011] and the projections of EPA [2012], as detailed in the supporting information.

Under this scenario, we find that HFC RF will plateau around 2035 and decline slightly by 2050 (Figure 1). By 2050, despite the global phase-down, HFC RF would be around 125% higher than 2012. However, when combined with the coincident reduction in CFC and HCFC RF, the proposed amendments to the Montreal

Protocol could result in a very significant climate impact, with a 26% decline in overall SGHG RF by 2050, compared to 2012, reaching 260 mW m^{-2} by 2050, and despite continued growth in FFGHG and HFC-23 RF.

The actual trajectory of future SGHG RF will depend critically on future emissions scenarios and their rate of removal from the atmosphere [e.g., Velders and Daniel, 2013]. To investigate the sensitivity of our projections to both of these factors, we ran the model forward using a suite of available projections and the currently accepted range of atmospheric lifetimes [SPARC, 2013]. We find that the single most important factor determining SGHG RF in 2050 will be the HFC emissions trajectory (Table 1). Therefore, we have examined the HFC phase-down schedule in contrast to a range of “no policy” HFC emissions trajectories (holding all other factors constant) to determine the possible influence of including HFCs in the Montreal Protocol (Figure 1).

The “HFC phase-down” trajectory leads to a 26% decline in RF by 2050, in contrast to an 11% to 42% increase in RF when we used scenarios from Gschrey *et al.* [2011] and Velders *et al.* [2009], or a more modest 13% decrease under RCP8.5. These differences equate to a $50\text{--}240 \text{ mW m}^{-2}$ reduction in global 2050 RF, equivalent to a cumulative emissions reduction of between 17 and 95 Pg $\text{CO}_2\text{-e}$ emissions, or 0.5–2.8 years of global CO_2 emissions, at 2010 levels [Boden *et al.*, 2013].

Our projections of SGHG emissions show that, if the proposed changes to the Montreal Protocol are implemented by 2050, the positive contributions to RF growth from SGHGs would be primarily due to the FFGHGs, SO_2F_2 , and HFC-23 (Figure 2). CFC-12 will still remain the dominant forcing agent and will still be responsible for most of the year-to-year reduction in RF. However, all HCFCs and all but the longest-lived HFCs (particularly HFC-23) could also contribute significantly to the reduction in SGHG RF.

Provided that suitable low-global warming potential alternatives to the HFCs can be found [e.g., Molina *et al.*, 2009; Daniel *et al.*, 2011; Velders *et al.*, 2012], which do not dramatically increase the energy intensity of the applications they are used in, the proposed changes to the Montreal Protocol would continue to make it a remarkably effective piece of global legislation for limiting the radiative impact of synthetic greenhouse gases.

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Erratum

In the originally published version of this article, four instances of text were incorrectly typeset due to typographical errors. The following have since been corrected and this version may be considered the authoritative version of record.

In section 3, Gg-CO₂e has been changed to Tg-CO₂e. In Figure 3, Gg yr⁻¹ has been changed to Tg yr⁻¹.